The Rate of Production of Helium from Radium.

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Some time ago I communicated a paper to the Society entitled "Note on the Use of the Radiometer in Observing Small Gas Pressures: Application to the Detection of the Gaseous Products produced by Radio-active Bodies."* In the course of the experiments recorded in that paper it was shown that a pressure of the fifty millionth of an atmosphere could easily be detected by radiometer motion, and that the helium produced by radio-active processes from some 10 milligrammes of bromide of radium could be definitely detected after a few hours. This led me to desire some direct measurements of the amount of helium produced by radium, and through the kindness of the Royal Society in allowing me the use of some radium chloride belonging to them I am able to give a condensed abstract of the experimental results so far obtained.

The salt employed was the 70 milligrammes of radium chloride prepared by Dr. T. E. Thorpe, F.R.S., for his determination of the atomic weight of radium, the preparation of which is fully described in 'Roy. Soc. Proc.,' A, vol. 80, p. 298.

The apparatus used for the measurements was a McLeod gauge in the construction of which no indiarubber joints were used; the mercury reservoir being connected to an exhaust pump, while the elevation and lowering of the mercury was carried out by admitting and exhausting air in the reservoir. The air coming in contact with the mercury was purified by passage over stick-potash and phosphoric anhydride. Sealed on to the gauge was a long U-tube containing a $\frac{1}{4}$ gramme of cocoanut charcoal placed in a small enlargement at the bend, the whole being arranged for liquid air or other cooling for any desired length of time. The object of the use of this cooled charcoal is to take up and condense all adventitious gases, other than hydrogen or helium, which might arise from minute leakage or otherwise be generated in the apparatus. The radium chloride was contained in a small bottle standing in a cylindrical glass bulb connected by a T-joint to the U-tube. To the other arm of the T was sealed a bulb containing about 15 grammes of cocoanut charcoal for producing a high exhaustion in the apparatus when cooled to $-190°$ C. The whole apparatus was well exhausted by mechanical

* 'Roy. Soc. Proc.,' A, vol. 79, p. 529, 1907.

means, all the glass tubes being heated as well as the charcoal receptacles and the radium chloride. On immersing the receptacle containing the 15 grammes charcoal in liquid air for some hours, while the $\frac{1}{4}$ gramme charcoal and the radium chloride were kept hot, an exhaust of 0·00015 mm. was obtained. This charcoal receptacle was now sealed off and the small $\frac{1}{4}$ gramme charcoal tube cooled in liquid air. In two hours an exhaust of 0·000054 mm. was reached.

The volume of the gauge and apparatus being approximately 200 c.c., a knowledge of the pressure in the apparatus gives by a simple calculation the actual volume of gas produced measured at atmospheric pressure and the temperature of the laboratory, and thus the rate of production of helium is obtained. This, referred to the weight of radium present, gives the increment in terms of cubic millimetres of gas per gramme of radium per day.

During the first three days the growth of pressure was very small, amounting to about 0·3 cub. mm. per gramme of radium per day. This was, however, practically all produced in the first day. On then heating the radium the pressure was increased to an amount corresponding to an increment of 0·99 cub. mm. The laboratory having to be closed for a fortnight no observations were taken, and as no part of the apparatus was cooled the emanation had free play throughout. About the 350th hour after heating the radium salt the pressure had increased to a value exactly corresponding to the 0·99 cub. mm. increment observed after the first three days. This rate was, however, only kept up for the two succeeding days during which the radium was not heated. On heating the radium a further increase of pressure was obtained (corresponding to a 1·1 cub. mm. increment measured from the start) which largely disappeared on cooling the radium and did not reappear on presently heating the radium again. From this stage throughout the next 120 hours the pressure rapidly became less, despite the heating of the radium, which only temporarily and partially restored it; and after 610 hours a lower pressure was recorded than that obtained immediately after the period of no observations, *i.e.* after 400 hours.

The charcoal was now heated to 450° C. by boiling sulphur. Then on again cooling it with liquid air the pressure was found to have been restored by an amount equal to one-third of that lost as stated above. This, however, quickly disappeared, and apart from fluctuations caused by heating, the radium remained during the next 300 hours at about the value observed just before heating the charcoal to 450° C.

At this point the charcoal was again heated in boiling sulphur and the previous result was repeated. In the ensuing 150 hours, however, the pressure, after falling a little, remained fairly steady, and then showed

a definite and maintained increase for three days, not permanently affected by again heating the charcoal to 450° C.

The quantity of permanent gas produced up to 1100 hours corresponded to an increment of 0·417 cub. mm. per gramme of radium per day taken over the whole period. At this point the radium was sealed off and the first experiment ended.

The large increase over the period in which the charcoal and the radium were both at ordinary temperatures may find some explanation from the unchecked action of the emanation on the charcoal, organic matter and combined moisture possibly present on the walls of the glass tubes of the apparatus, in quantities though small yet large enough to produce the total amount of gas present which, as measured with charcoal at the ordinary temperature, corresponded to a pressure of 1—2 mm.

It may be noted that on the supposition of this gas being largely due to a continuous air leak, the amount of uncondensed gases of the helium type thus introduced would be infinitesimal.

Apart from the possible presence in the apparatus of organic material the radium itself might conceivably at the beginning have been contaminated with traces of organic matter, and a further experiment was decided on to which these objections could not be applied.

In the second experiment the gauge as well as the connecting tubes were well cleaned out with nitric acid and all thoroughly dried. The radium, after the 1100 hours in which it was under high exhaustion and had been frequently heated, was certainly in a more satisfactory condition. Further, to prevent the unchecked action of the emanation throughout the apparatus, the little charcoal condenser was maintained at a degree or two below that of the boiling point of oxygen by the use of old liquid air for a period of about six weeks. A larger quantity of charcoal was used, viz., 1 gramme, the more effectively to condense out extraneous gases while leaving any helium substantially unaffected. This charcoal had been treated with chlorine at a red heat and subsequently with hydrogen.

Beyond this the conduct of the experiment followed the lines of the former one. The mercury pump exhaust was continued for several hours and was carried to 0·002 mm. The large charcoal bulb was then cooled for several hours in liquid air while heating the 1 gramme of charcoal and the radium salt. A pressure as low as 0·00005 mm. was thus obtained when the charcoal was sealed off. On now placing the **U**-tube containing a small quantity of charcoal in liquid air the pressure registered was 0·000044 mm.

These conditions were maintained for five days, during which a steady

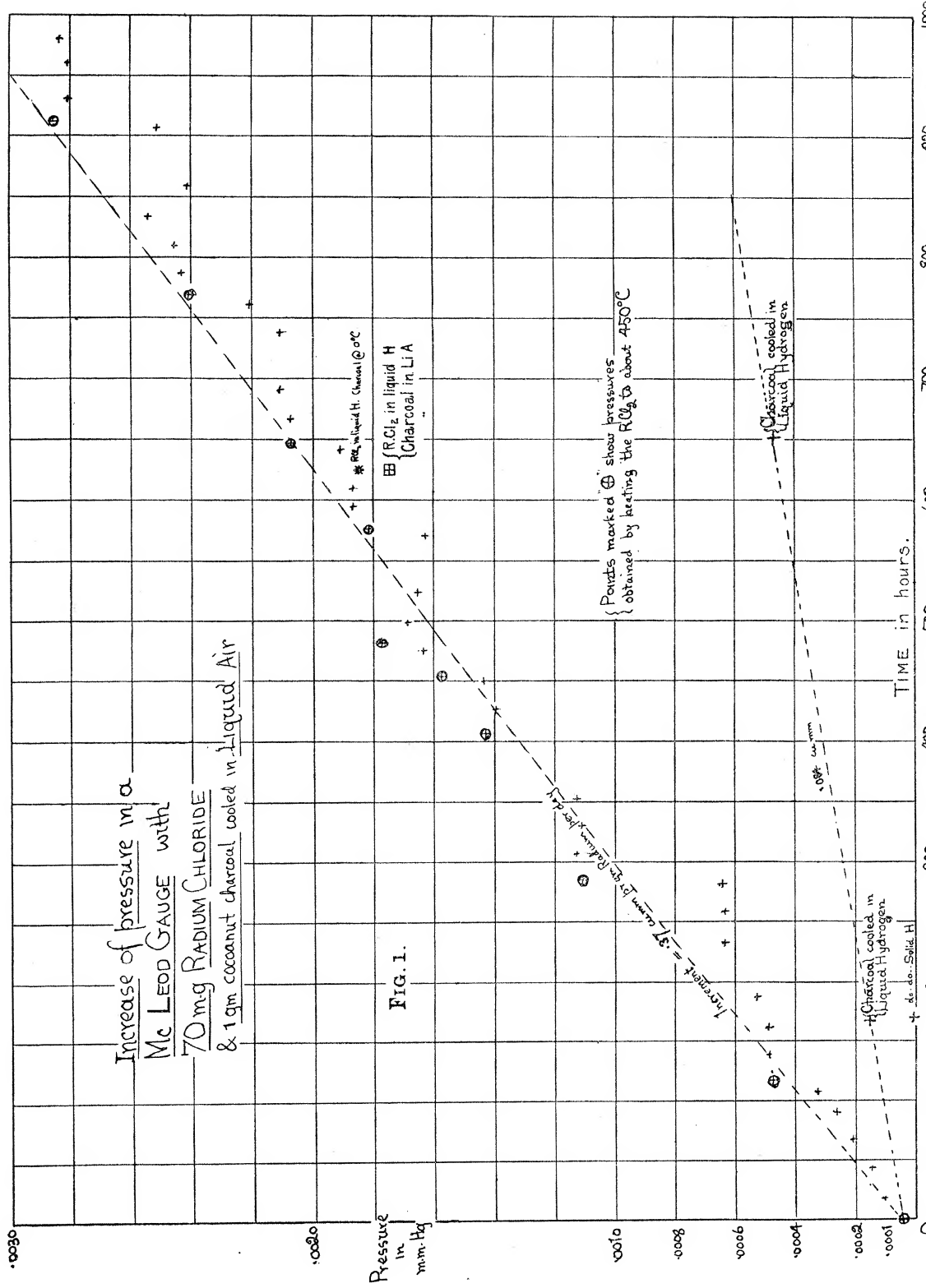
growth of pressure was observed corresponding to an increment of approximately 0·3 cub. mm. per gramme of radium per day. The radium was then heated with a small Bunsen flame as before to a low red heat, when the pressure was increased by about 40 per cent. This increase showed no sign of disappearing, but during the next week a decided but somewhat irregular growth of pressure was recorded. The radium was again heated, when a further increase of pressure was observed. In the succeeding five days it remained steady, only to be again increased on heating the radium. This treatment was repeated in all 10 times at varying intervals during 1100 hours, and in each case the pressure rose on heating and remained fairly steady on standing. All the observations of the second set of experiments are graphically represented in fig. 1. A mean line is drawn through the observations taken with the radium heated, giving a steadily maintained helium increment of approximately 0·37 of a cub. mm. per gramme of radium per day.

In order to ascertain if any helium was occluded in the cooled charcoal and the surrounding glass, the latter was raised to a low red heat while the tube containing the radium chloride was temporarily cooled in liquid air, with the object of condensing out and localising the emanation coming from the heated charcoal and preventing its access to the gauge. The temperature was maintained for an hour, and then the charcoal was allowed to cool and finally replaced in the liquid air. The radium chloride was then allowed to warm up and was heated to near a low red for a short time. After these alternations no increase in pressure was observed, from which it may be inferred that the occlusion of the helium takes place mainly in that part of the apparatus where the radium chloride is situated.

On two occasions the charcoal was cooled in liquid hydrogen, viz., after 165 hours, and again after 650 hours. The proportionate reduction of pressure was the same in both cases, tending to show that the composition or nature of the gas remaining uncondensed by the liquid air remained the same throughout, although steadily increasing in quantity.

In reference to this last point a separate experiment was made in which pure helium under a small tension, produced by heating 0·5 gramme uranite and passing the gas produced over 1 gramme charcoal cooled in liquid air, was subjected to the action of $\frac{1}{4}$ gramme of clean exhausted charcoal at the temperature of liquid air and liquid hydrogen respectively. The ratio of the two pressures so obtained was in close agreement with that observed in the radium experiment.

A further test of the purity of the gas producing the permanent pressure observed in the radium experiment with the charcoal cooled in liquid air was made by simply cooling the bulb containing the radium in liquid


Increase of pressure in a
Mc Leod Gauge with
70 mg Radium Chloride
& 1 gm coconut charcoal cooled in Liquid Air
Pressure in mm. Hg
Time in hours.
.0030
.0020
.0010
.0008
.0006
.0004
.0002
.0001
0
50
100
200
300
400
500
600
700
800
900
1000
⊞ {R.Cl2 in liquid H
{Charcoal in Li A
{Points marked ⊕ show pressures
{obtained by heating the RCl2 to about 450°C
Increment = 3.7 mm. per Radium per day
+Charcoal cooled in Liquid Hydrogen
+Charcoal cooled in Liquid Hydrogen
+ do. do. Solid H
FIG. 1.

hydrogen, allowing the charcoal meanwhile to warm up to 0° C. If any hydrogen was present in the gas it is certain that there would be an increase of pressure recorded, since although hydrogen is partially absorbed by charcoal in liquid air yet it would not be reduced in pressure by cooling in liquid hydrogen. On allowing the charcoal therefore to warm up, any hydrogen expelled would remain and cause an increased pressure. Inasmuch as an increase was not recorded, it can be safely assumed that no hydrogen is present, and thus the gas pressure measured consists entirely of helium.

A confirmation of this was made spectroscopically as follows:—Two tin-foil electrodes were placed round the narrow capillary measuring tube of the gauge, near the closed end. These were about 3 cm. long and about $1\frac{1}{2}$ cm. apart and were wired on with thin copper wire. The gas was compressed into this capillary space, as in taking an ordinary measure to any pressure of the order of 2 or 3 mm., while an induction discharge passed in the gas. The spectroscopic examination of this discharge revealed only the six principal helium lines, mercury, and a trace of the carbonic oxide spectrum. I have shown that the carbonic oxide spectrum always occurs in electrode-less tubes.*

I am not aware of any previous direct measurements of the rate of production of helium from radium, but in a paper on "Some Properties of Radium Emanation," by A. J. Cameron and Sir William Ramsay,† the ratio of the amount of helium produced to that of the emanation was found to be 3·18, and as the amount of the emanation found by them was about 1 cub. mm. per gramme of radium per day, the resulting helium according to this experiment ought to reach about 3 cub. mm. or at least eight times the rate of production found in the above experiments. I am at a loss to explain the origin of such grave discrepancies in the measured amount of the helium produced by radium.‡ On the other hand, Professor Rutherford, in his work entitled 'Radio-active Transformations,' 1906, p. 186, on the theoretical assumption that the α-particle is an atom of helium carrying twice the ionic charge, deduced from electrical measurements that the number of particles expelled per year per gramme of radium would reach 4×10^{18}, and as 1 c.c. of a gas at standard temperature and pressure contains $3{\cdot}6 \times 10^{19}$ molecules, the volume of helium produced per year would amount to 0·11 c.c., which

* See paper ('Roy. Soc. Proc.,' vol. 64, p. 237) "On the Application of Liquid Hydrogen to the Production of High Vacua; together with their Spectroscopic Examination."

† 'Chem. Soc. Jour.,' 1907, p. 1274.

‡ Professor Rutherford, in a paper, "Experiments with Radium Emanation," 'Phil. Mag.,' July, 1908, shows this result is at least ten times too great, his value being of the order 0·11 cub. mm. of emanation per day, whereas from my experiments the rate of helium production is just three times this amount.

is equivalent to about 0·3 of a cub. mm. per day. Considering I have found a rate of helium production of the order of 0·37 cub. mm., the agreement between experiment and the theoretical prophecy of Rutherford is almost too wonderful, substantiating as it does the accuracy of the theory of radio-active changes he has done so much to initiate and develop.

I have to express my obligations to Mr. Robert Lennox, F.C.S., and Mr. W. J. Green, B.Sc., for aid given in the conduct of these long and laborious experiments.

On the Reflection of Waves from a Stratum of Gradually Varying Properties, with Application to Sound.

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In a variable medium, the velocity of propagation of a train of waves, and the wave-length at any point, are functions of the position of that point. The circumstances of such a propagation have only been worked out in detail in one particular case. Lord Rayleigh,* in connection with the transverse vibrations of a string of variable density, dealt very completely with the case in which the density is inversely proportional to the distance from a fixed point. In his original investigation† the results were applied to the corresponding optical problem, and a numerical example given.

Although this is perhaps the only interesting case in which a simple exact solution appears possible, yet a close approximation may be made to the existing conditions, even in the general problem, when the waves are short in comparison with the other distances concerned. The development of such a theory, with an examination of some important cases, is the object of the present paper.

Let V_0 be the velocity of a plane wave-train at some point of a medium, which we may choose as origin, and suppose that the train is advancing along the direction x.

* 'Theory of Sound,' vol. 1, § 148.

† 'Proc. Lond. Math. Soc.,' vol. 11, 1880, pp. 51—56; 'Collected Papers,' vol. 1, pp. 460—465.